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SCIENTIFIC REPORT  
A: SORPTION BY CO<sub>2</sub> BETWEEN 7125 AND 8000 cm<sup>-1</sup>  
(1.25-1.40 Microns)

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# ABSTRACT

Transmission spectra in the  $7125\text{-}8000\text{ cm}^{-1}$  region have been obtained for  $\text{CO}_2$  samples with absorber thicknesses as great as  $2.18 \times 10^5\text{ atm cm}_{\text{STP}}$ . Three absorption bands not observed previously have been identified, and the strengths of several bands have been determined. Transmission spectra and a table of integrated absorptance versus wavenumber are included for five representative samples.

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## INTRODUCTION AND SUMMARY

The region between 7125 and 8000  $\text{cm}^{-1}$  contains several  $\text{CO}_2$  absorption bands which are too weak to play a very important part in the radiative transfer in the earth's atmosphere. However, since a large portion of this region contains very little absorption by  $\text{H}_2\text{O}$  and other gases in the earth's atmosphere,  $\text{CO}_2$  absorption by the atmospheres of other planets can be studied at ground level with little interference from our own atmosphere. In order to interpret the results of such a study, it is necessary to identify the bands and to know the strengths, widths, and positions of absorption lines.

Courtoy<sup>1</sup> has observed and identified the more important bands in this region and has determined the positions of many of the lines. We have used larger samples in order to study the weaker bands and have determined the strengths of several bands from measurements on samples at high pressures. Besides a listing of the strengths of several of the bands, this report contains detailed results for five representative samples. Included are replotted spectra with several of the bands identified and a table of integrated absorptance versus wavenumber. Tables of transmittance versus wavenumber are available from the authors for workers who require them.

## EXPERIMENTAL

Samples of  $\text{CO}_2$  were contained in two different multiple-pass absorption cells. The longer one was used at path lengths of 469 and 933 meters and at 1 and 2.5 atm pressure. The shorter one was employed at 14.6 atm with 16.5 and 32.9 meter paths. The absorption cells and sampling procedure have been described previously in considerable detail.<sup>2,3</sup>

The absorber thickness  $u$  was calculated by use of the following equation.

$$u(\text{atm cm}_{\text{STP}}) = p(\text{atm}) L(\text{cm}) [1 + 0.005p]^{273/296}, \quad (1)$$

where  $p$  is the  $\text{CO}_2$  pressure and  $L$  is the geometrical path length. The factor  $(273/296)$  accounts for the difference in densities at standard temperature ( $273^\circ\text{K}$ ) and at room temperature ( $296^\circ\text{K}$ ) at which the measurements were made. The quantity  $(1 + 0.005p)$  accounts for the non-linearity in the relation between the density of  $\text{CO}_2$  and its pressure.

The  $\text{CO}_2$  contained traces of  $\text{H}_2\text{O}$  impurity which caused absorption near a few of the  $\text{CO}_2$  bands. No absorption by other impurities was observed. It is probably safe to assume that all the isotopes were present in their natural abundances ( $\text{C}^{12}$ , 98.9%;  $\text{C}^{13}$ , 1.1%;  $\text{O}^{16}$ , 99.76%;  $\text{O}^{17}$ , 0.04%;  $\text{O}^{18}$ , 0.20%).

Spectra were scanned by the use of a small grating spectrometer enclosed in a vacuum tank. The transmittance of a sample was determined by comparing its spectrum with a background curve scanned over the same region and with the same experimental conditions except that the absorption cell was evacuated. Each spectrum was examined and compared with others as a check for consistency. Spectra of  $\text{H}_2\text{O} + \text{N}_2$  were used to determine the contribution of the  $\text{H}_2\text{O}$  impurity. Each spectrum was then replotted and digitized by the method described previously.<sup>2,3</sup> Pairs of values related to transmittance and wavenumber were punched on IBM cards which served as



input for a computer program used to calculate transmittance and integrated absorptance as a function of wavenumber. The sample parameters for five representative samples scanned with a spectral slitwidth of approximately  $1.3 \text{ cm}^{-1}$  are given in Table 1. The positions of many of the  $\text{CO}_2$  lines given by Courtoy<sup>1</sup> and  $\text{H}_2\text{O}$  lines from an article by Nelson<sup>4</sup> were used for wavenumber calibration.

TABLE 1  
SAMPLE PARAMETERS

Sample No.	p torr	p atm	u atm $\text{cm}_{\text{STP}}$	L Path Meters
1	1,900	2.50	$2.18 \times 10^5$	933
2	1,900	2.50	$1.09 \times 10^5$	469
3	760	1.00	$8.65 \times 10^4$	933
4	11,100	14.6	$4.76 \times 10^4$	32.9
5	11,100	14.6	$2.38 \times 10^4$	16.5

## RESULTS AND DISCUSSION

Table 2 contains a list of all the  $\text{CO}_2$  bands in the  $7125\text{-}8000\text{ cm}^{-1}$  region which one might expect to absorb appreciably for the samples included in the present investigation. The positions of the centers of several of the bands have been measured by Courtoy.<sup>1</sup> The centers of the other bands were determined from energy levels calculated by Stull, Wyatt, and Plass.<sup>5</sup>

Since, for  $\text{CO}_2$ ,  $v_1 \cong 2v_2$  ( $1388.2 \cong 2 \times 667.4$ ),<sup>1</sup> bands arising from transitions for which  $\Delta v_3$  is the same and  $2\Delta v_1 + \Delta v_2$  is constant occur near each other. ( $\Delta v_1$  is the change in  $v_1$ , the quantum number associated with  $v_1$ , etc.) Most of the absorption in the  $7125\text{-}8000\text{ cm}^{-1}$  region is due to bands in which  $\Delta v_3 = 1$ , and  $2\Delta v_1 + \Delta v_2 = 8$ . The first five bands listed in Table 2 form a group in which  $\Delta v_1$  varies from 0 to 4, with  $\Delta v_2$  taking on even values between 0 and 8. The lower energy level involved in the transitions for these five bands is  $00^0_0$ .

In any theoretical analysis of such a group of bands, it makes little difference which one of the sets of quantum numbers corresponds to a given band. In fact, because of Fermi interactions, it is impossible in some instances to be sure of the proper assignment. For this reason, it has been suggested recently that a new notation be adopted for  $\text{CO}_2$  bands. Although the new notation used in this report may be slightly different from the one which eventually will have the most widespread usage, it is certainly similar in some respects. In the new notation, all the energy levels in a group are denoted by three numbers in the following order: the maximum value of  $v_1$ , the minimum  $v_2$ , and  $v_3$ . These three numbers are followed by Roman numeral subscripts I, II, . . ., with I assigned to the highest level, II to the second highest, etc. Table 2 includes both the old and new notations for the energy levels involved in the transitions.

TABLE 2  
CO<sub>2</sub> ABSORPTION BANDS BETWEEN 7125 AND 8000 cm<sup>-1</sup>

Band Center cm <sup>-1</sup>	Authority <sup>a</sup>	Upper Level <sup>b</sup>		Remark
		Old Notation	New Notation	
7920.5	SWP	40 <sup>0</sup> <sub>1</sub>	401 <sub>I</sub>	
7734.30	C	32 <sup>0</sup> <sub>1</sub>	401 <sub>II</sub>	
7593.54	C	24 <sup>0</sup> <sub>1</sub>	401 <sub>III</sub>	
7460.37	C	16 <sup>0</sup> <sub>1</sub>	401 <sub>IV</sub>	
7284.22	SWP	08 <sup>0</sup> <sub>1</sub>	401 <sub>V</sub>	
7592.44	C	32 <sup>0</sup> <sub>1</sub>	401 <sub>II</sub>	C <sup>13</sup> <sub>2</sub> <sup>16</sup> <sub>2</sub> <sup>c</sup>
7481.34	C	24 <sup>0</sup> <sub>1</sub>	401 <sub>III</sub>	C <sup>13</sup> <sub>2</sub> <sup>16</sup> <sub>2</sub>
7332.39	SWP	16 <sup>0</sup> <sub>1</sub>	401 <sub>IV</sub>	C <sup>13</sup> <sub>2</sub> <sup>16</sup> <sub>2</sub>
7758.3	SWP	33 <sup>1</sup> <sub>1</sub>	411 <sub>II</sub>	Lower Level is 01 <sup>1</sup> <sub>0</sub> (010 <sub>I</sub> )
7584.4	SWP	25 <sup>1</sup> <sub>1</sub>	411 <sub>III</sub>	Lower Level is 01 <sup>1</sup> <sub>0</sub> (010 <sub>I</sub> )
7415.2	SWP	17 <sup>1</sup> <sub>1</sub>	411 <sub>IV</sub>	Lower Level is 01 <sup>1</sup> <sub>0</sub> (010 <sub>I</sub> )
7901.5	SWP	13 <sup>1</sup> <sub>2</sub>	212 <sub>II</sub>	
7743.8	SWP	05 <sup>1</sup> <sub>2</sub>	212 <sub>III</sub>	
7981.17	C	02 <sup>0</sup> <sub>3</sub>	103 <sub>II</sub>	C <sup>13</sup> <sub>2</sub> <sup>16</sup> <sub>2</sub>

<sup>a</sup> Authority for position of band center. C denotes experimental value from Courtoy.<sup>1</sup> SWP indicates that the band center has been determined from energy levels calculated by Stull, Wyatt, and Plass.<sup>5</sup>

<sup>b</sup> The upper level of the transition is given in both the old and new notation. The lower level is 00<sup>0</sup><sub>0</sub> (000<sub>I</sub>) unless indicated otherwise.

<sup>c</sup> Molecular species is C<sup>12</sup><sub>2</sub><sup>16</sup><sub>2</sub> except as indicated.

Transmittance spectra of the five samples listed in Table 1 are shown in Fig. 1 with the corresponding sample numbers enclosed in squares. Most of the absorption between 7400 and 7800  $\text{cm}^{-1}$  is due to the  $401_{\text{II}}$ ,  $401_{\text{III}}$ , and  $401_{\text{IV}}$  bands of the most abundant isotopic species,  $\text{C}^{12}\text{O}_2^{16}$ . The corresponding bands for the  $\text{C}^{13}\text{O}_2^{16}$  isotope cannot be seen in our results; the  $401_{\text{II}}$  and  $401_{\text{III}}$  bands are masked by stronger bands which overlap them, and the  $401_{\text{IV}}$  band, whose band center is calculated to be at 7332.39  $\text{cm}^{-1}$ , was not observed since it is probably very weak and occurs near some  $\text{H}_2\text{O}$  lines appearing in the spectrum as a result of the impurity in the sample. Absorption by the less abundant isotopes and by weaker bands of  $\text{C}^{13}\text{O}_2^{16}$  would, of course, be less and probably not observable in the spectra of samples included in the present investigation.

Evidence of the difference band  $411_{\text{II}} \leftarrow 010_{\text{I}}$  can be seen on the high wavenumber side of its associated combination band  $401_{\text{II}}$  which occurs 24  $\text{cm}^{-1}$  below. (The change in all the quantum numbers are the same for a combination band as for its associated difference band.) The  $411_{\text{III}} \leftarrow 010_{\text{I}}$  band is masked by its associated combination band  $401_{\text{III}}$ , while slight evidence of the  $411_{\text{IV}} \leftarrow 010_{\text{I}}$  band appears just above 7400  $\text{cm}^{-1}$  on the low wavenumber side of  $411_{\text{IV}}$ . A few  $\text{H}_2\text{O}$  lines near 7400  $\text{cm}^{-1}$  make it difficult to determine the true shape of the  $\text{CO}_2$  spectrum in this region. The  $212_{\text{III}}$  band whose center is at 7743.8  $\text{cm}^{-1}$  is probably masked by the stronger  $401_{\text{II}}$  band.

The bands near 8000  $\text{cm}^{-1}$  and the one near 7300  $\text{cm}^{-1}$  are so weak that only the sample having maximum absorber thickness (Sample 1) produced appreciable absorption. There is considerable uncertainty in the spectrum near 7300  $\text{cm}^{-1}$  because of possible errors in accounting for absorption by the  $\text{H}_2\text{O}$  impurity. Because of the small amount of absorption in the 7850-8000  $\text{cm}^{-1}$  region, the true shape of the spectrum is difficult to determine; the curve of this region in Fig. 1 represents the average of three curves. The transmittance minimum near 7900  $\text{cm}^{-1}$  is probably due to the Q-branch of the  $212_{\text{II}}$  band, and the two maxima near 7920 and 7980 are probably at the centers of the bands indicated. No absorption other than that shown in Fig. 1 was observed between 7125 and 8000  $\text{cm}^{-1}$ .

The  $212_{\text{I}}$  band and the remainder of the 103 group for both  $\text{C}^{13}\text{O}_2^{16}$  and  $\text{C}^{12}\text{O}_2^{18}$  occur at higher wavenumbers and have been discussed previously.<sup>6</sup>

(13)

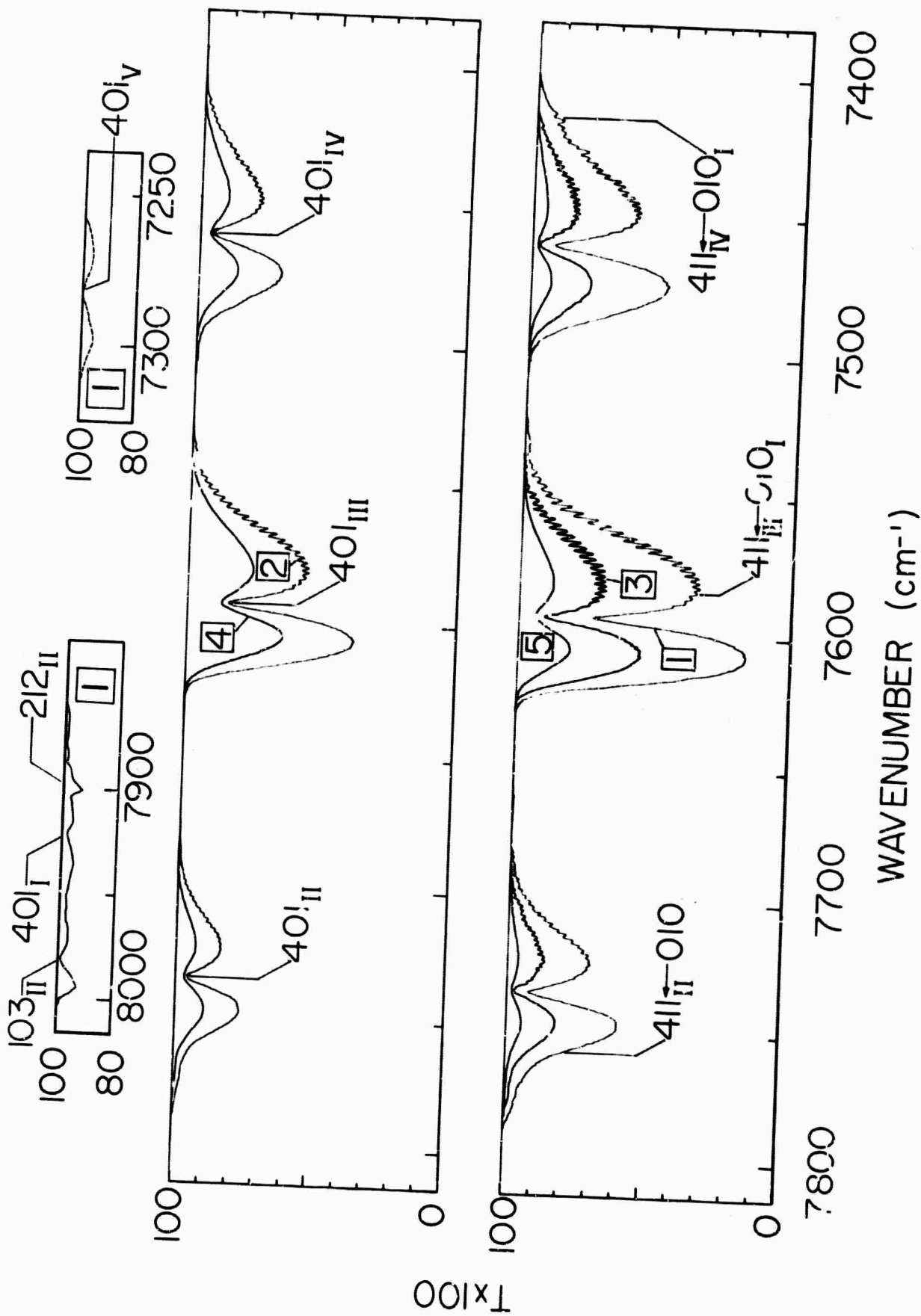


FIG. 1 TRANSMISSION SPECTRA OF CO<sub>2</sub> BETWEEN 7250 AND 8000 cm<sup>-1</sup>.

No absorption other than that indicated was observed between 7125 and 8000 cm<sup>-1</sup>. The sample numbers are enclosed in squares, and the positions of several band centers are indicated. A Cl<sub>3</sub>O<sub>2</sub>Cl<sub>6</sub> band is denoted by (13); all others are Cl<sub>2</sub>O<sub>2</sub>Cl<sub>6</sub>.

## INTEGRATED ABSORPTANCE

Values of the integrated absorptance  $\int_{\nu'}^{\nu} A(\nu) d\nu$  are presented in Table 3 for Samples 1-5. The three parts of the table correspond to the three stronger regions of absorption between 7400 and 7800  $\text{cm}^{-1}$ . Each column represents the sample indicated at the top. The lower limit of integration,  $\nu'$ , which is shown at the top of each column, was chosen at a point where there was no absorption. The integrated absorptance between any two wavenumbers listed can be found by subtracting the values tabulated at those two points. In regions where there is structure in the recorded spectra, the integral is tabulated at points midway between the centers of the absorption lines. In other regions where the spectra are smooth, tabulations are made in intervals of 2 or 5  $\text{cm}^{-1}$ .

Table 3  $\int_{\vec{v}} A(\vec{v}) d\vec{v}$ [illegible]

## BAND STRENGTHS

The strength, or intensity, of an absorption band is given by

$$S_v = \int K(v) dv, \quad (2)$$

where the integration is performed over all  $v$  for which there is appreciable absorption by the band.  $K(v)$ , the absorption coefficient, is related to  $T'(v)$  the true transmittance that would be observed with infinite resolving power by

$$K(v) = -\frac{1}{u} \ln T'(v). \quad (3)$$

In regions where two or more bands overlap each other, the value of  $K(v)$  used in (2) to determine a band strength must include only the contribution by the band of interest. We have shown in a previous report<sup>7</sup> that under conditions of sufficiently high pressure and high transmittance,  $\int \ln T'(v) dv \approx \int \ln T(v) dv$ , where  $T(v)$  is the observed transmittance. Thus it is possible to determine the strength of a band, or at least of a region containing overlapping bands, from the spectrum of a sample satisfying these conditions.

From the transmission spectra shown in Fig. 1, we were able to determine  $-\frac{1}{u} \int \ln T(v) dv$  for different spectral regions. We then estimated the contributions of some of the weaker bands in order to calculate the strengths of overlapping stronger bands. The results are summarized in Table 4. For example,  $-\frac{1}{u} \int \ln T(v) dv = 12.2 \times 10^{-5} \text{ atm}^{-1} \text{ cm}^{-1} \text{ STP cm}^{-1}$  for the 7400-7500  $\text{cm}^{-1}$  region. We know that at least three bands contribute to the absorption in this region: the  $401_{IV}$  and its associated difference band  $411_{IV} \leftarrow 010_I$ , as well as the  $401_{III}$  band of  $C^{14}_{16}$ . The contribution of the latter band is very small ( $\sim 0.3 \times 10^{-5}$ ) and can be estimated from the strength of the corresponding band of  $C^{12}_{16}$  by assuming that their strengths are proportional to the abundances of the C isotopes (1.1/98.9). We can also show from quantum theory<sup>8</sup> that, at room temperature, the strength of a  $\Pi \leftarrow \Pi$  difference band such as  $411_{IV} \leftarrow 010_I$  is 0.078 times that of the associated combination band  $401_{IV}$  which arises from a transition from the  $000_I$  state. By using this relationship, we were able to show that the strength of the  $401_{IV}$  band is  $11.0 \times 10^{-5} \text{ atm}^{-1} \text{ cm}^{-1} \text{ STP cm}^{-1}$ . A similar method was used for the  $401_{III}$  band. In order to estimate the strength of the  $401_{II}$  band, we had to assume a value for the  $212_{III}$  band. For lack of anything better we used  $0.45 \text{ atm}^{-1} \text{ cm}^{-1} \text{ STP cm}^{-1}$ , the value determined for the



TABLE 4  
STRENGTHS OF ABSORPTION BANDS

Band Center cm <sup>-1</sup>	Upper Level	$S_v^*$ atm <sup>-1</sup> cm <sup>-1</sup> STP cm <sup>-1</sup>	
7920.5	401 <sub>I</sub>	0.55 ± 0.25 × 10 <sup>-5</sup>	
7734.30	401 <sub>II</sub>	7.65 ± 0.6 × 10 <sup>-5</sup>	Based on 8.75 × 10 <sup>-5</sup> for 7675-7790 cm <sup>-1</sup> region
7593.54	401 <sub>III</sub>	28.6 ± 1.5 × 10 <sup>-5</sup>	Based on 31.1 × 10 <sup>-5</sup> for 7510-7630 cm <sup>-1</sup> region
7460.37	401 <sub>IV</sub>	11.0 ± 0.6 × 10 <sup>-5</sup>	Based on 12.2 × 10 <sup>-5</sup> for 7400-7500 cm <sup>-1</sup> region
7284.22	401 <sub>V</sub>	0.55 ± 0.25 × 10 <sup>-5</sup>	
7901.5	212 <sub>II</sub>	0.45 ± 0.20 × 10 <sup>-5</sup>	
7981.17	103 <sub>II</sub> C <sup>13</sup> O <sup>16</sup> <sub>2</sub>	0.60 ± 0.20 × 10 <sup>-5</sup>	

\* The subscript (STP) is used to emphasize that the strengths were calculated by using absorber thicknesses normalized to standard temperature and pressure. The measurements were made at room temperature 296°K.

212<sub>II</sub> band. If the actual strength is less than our assumed value, only a few percent error is introduced in our value for the 401<sub>II</sub> band. It seems unlikely that the 212<sub>III</sub> band is significantly stronger than we assumed since there is no strong evidence for it in the spectra shown in Fig. 1. Furthermore, Courtois<sup>1</sup> did not observe it in his high-resolution spectra.

Although there are no overlapping CO<sub>2</sub> bands, the uncertainty in the strength of the 401<sub>V</sub> band is large because of errors in accounting for H<sub>2</sub>O absorption. Because of the small amount of absorption and overlapping of the bands between 7850 and 8000 cm<sup>-1</sup>, the strengths of the bands in this region could not be determined accurately. The contribution of each band was estimated by dividing the absorption into portions which seemed consistent with the contour of the spectrum shown in Fig. 1.

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13. **ABSTRACT:** Enter an abstract giving a brief and factual summary of the document indicative of the report, even though it may also appear elsewhere in the body of the technical report. If additional space is required, a continuation sheet shall be attached.

It is highly desirable that the abstract of classified reports be unclassified. Each paragraph of the abstract shall end with an indication of the military security classification of the information in the paragraph, represented as (TS), (S), (C), or (U).

There is no limitation on the length of the abstract. However, the suggested length is from 150 to 225 words.

14. **KEY WORDS:** Key words are technically meaningful terms or short phrases that characterize a report and may be used as index entries for cataloging the report. Key words must be selected so that no security classification is required. Identifiers, such as equipment model designation, trade name, military project code name, geographic location, may be used as key words but will be followed by an indication of technical context. The assignment of links, rules, and weights is optional.